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(54) Title: FOAMED BLENDS OF PROPYLENE-BASED POLYMERS

(57) Abstract: Foamable polymer blends, foamed polymer compositions made therefrom, and processes for making such blends and foamed compositions, are disclosed. The polymer blends include at least a first polymer component, including a propylene homopolymer or a copolymer of propylene and a minor amount (less than 10 % by weight) of ethylene or an alpha-olefin, and a second polymer component, including one or more random copolymers of propylene (65 to 95 % by weight) and ethylene or an alpha-olefin. The polymer blends contain from 50 % to 99.9 % by weight of the first polymer component, and have a crystallinity of from 2 % to 65 % from isotactic propylene sequences. Physical or chemical blowing agents are used to foam the polymer blend, to form a foamed polymer blend of the present invention having improved elastic properties.



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FOAMED BLENDS OF PROPYLENE-BASED POLYMERS

FIELD OF THE INVENTION

The present invention relates generally to foamed polymers, and more specifically, to foamed blends of polymers having improved properties. In particular, the present invention provides foamed blends of propylene-based polymers having improved elastic properties.

BACKGROUND

Polypropylene is one of the most widely used and versatile polymers, finding applications in oriented and non-oriented films, textile fibers, nonwoven fibers, and injection molded parts, for example. Compared to other commonly used polymers, such as polyethylene and polystyrene, polypropylene has several advantages. For example, polypropylene has a higher modulus and heat deflection temperature than polyethylene, making it better suited to durable applications in the appliance and automotive segments. Compared to polystyrene, polypropylene is preferred in applications requiring good organoleptic performance and high barrier properties. In addition, polypropylene has good resistance to both acids and bases, and good blending ability with a variety of other polymers.

Although polypropylene is widely used in many applications, it has not been as successfully used in applications requiring foamed polymers. Foamed polymers are used in automotive, marine, and appliance and packaging applications because of their good insulating and structural properties at low added weight. Polypropylene-based foams are distinguished from other foams in their ability to withstand higher temperatures, a feature of particular utility in automotive applications. Generally, however, polypropylene foams are not very elastic, and their utility is thereby limited.

The deficiencies in polypropylene foams are believed to be related to the generally poor melt strength and rapid melt viscosity drop, poor sheet sag and comparatively slow crystallization kinetics of polypropylene. For example, to successfully foam an article formed from a polyolefin, it is desirable that the

polyolefin have a high melt strength. With high melt strength, the bubble growth rate within the polyolefin can be controlled without premature bursting. Controlling bubble growth rate is also important to ensure a uniform distribution of cell sizes, which leads to greater product uniformity. Additionally, broader
5 polymer processing temperature windows are desirable so that when the polymer is used in an article forming process, temperature variances along the process line are less disruptive to the fabrication of a high quality product. Unfortunately, the disadvantageous properties of conventional polypropylenes have made development of such foams and articles made therefrom difficult.

10 WO 97/40080 discloses elastomer-modified polypropylene compositions which are said to be suitable for foaming, among numerous other potential applications. The elastomer-modified polypropylene composition is formed from: (a) 20-80% by weight of a very high molecular weight random copolymer of propylene and ethylene; (b) 80-20% by weight of an essentially homopolymeric
15 low molecular weight polypropylene; and (c) 5 to 40% by weight of a "rubbery copolymer". Reportedly, component (a) provides extremely high shape resistance; component (b) provides good melt-processing properties and improved stiffness; and component (c) provides better impact resistance. The composition is produced using a catalyst system which includes the reaction product of a
20 tetravalent titanium compound and a magnesium halide compound, which is known to produce copolymers with a broad molecular weight distribution and non-uniform composition distribution.

EP 0 633 289 A1 discloses foamable blends of 5-50% of a crystalline propylene homopolymer or copolymer and 50-95% of an elastomeric polymer of
25 ethylene, propylene and/or a C₄-C₁₀ α -olefin. The composition is produced using a Ziegler-Natta catalyst system known to produce a broad compositional distribution. In addition, the composition is partially cross-linked.

EP 0 472 946 B1 discloses a composition including: (a) 10-50 parts by weight of propylene or an ethylene-propylene copolymer, the homopolymer or
30 copolymer having an isotactic index greater than 80; (b) 5-20 parts by weight of an ethylene-containing copolymer fraction insoluble in xylene; and (c) 40-80 parts by weight of a copolymer of ethylene and propylene or a C₄-C₁₀ α -olefin, the

copolymer being soluble in xylene and having an intrinsic viscosity of from 1.5 to 4 dl/g. The composition is produced using a magnesium chloride-titanium chloride reaction product catalyst.

It would be advantageous to develop polypropylene compositions with a balance of properties better suited to foam applications. In particular, it would be advantageous to develop foamed polypropylene compositions having good heat resistance as well as good elastic properties, such as high elongation, good recovery, and low compression set.

For additional background, see also: USPN 6,040,348; USPN 4,870,114; USPN 4,832,770; USPN 4,766,159; USPN 3,876,494; USPN 3,846,349; WO 97/40080; and EP 633,289.

SUMMARY OF THE INVENTION

The present invention provides foamable polymer blends, and foamed polymer compositions made therefrom, having improved properties. In one embodiment, the polymer blends include at least two polymer components. The first polymer component ("FPC") can be a propylene homopolymer or a copolymer of propylene and a minor amount (less than 10% by weight) of ethylene or an alpha-olefin. The second polymer component ("SPC") includes one or more random copolymers of propylene (65 to 95% by weight) and ethylene or an alpha-olefin, and has a crystallinity of from 2% to 65% from isotactic propylene sequences. The blend contains from 50% to 99.9% by weight of the first polymer component, based on the total weight of the first and second polymer components. Preferably, the first polymer component has a melting point greater than 110°C, and the second polymer component has a melting point of from 25°C to 105°C. Physical or chemical blowing agents are used to foam the polymer blend, to form a foamed polymer blend of the present invention.

In another embodiment, the polymer blends include at least two polymer components, a first polymer component as described above, and a second polymer component including one or more random copolymers of propylene (65 to 95% by weight) and ethylene.

In another embodiment, the present invention is directed to a process for preparing a foamed polymer composition, the process comprising the steps of forming any of the polymer blends described herein, and contacting the polymer blend with a chemical or physical foaming agent sufficient to produce a foamed polymer composition.

In still another embodiment, the present invention is directed to articles of manufacture formed from or including any of the foamed polymer blends of the invention.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 shows the elongation performance of polymer blends used for the foams of the present invention.

Figure 2 shows the elongation performance of prior art polymeric materials.

Figure 3 shows the flexural modulus of polymer blends used for the foams of the present invention.

Figure 4 shows the flexural modulus performance of prior art polymeric materials.

Figures 5A and 5B show electron micrographs of a prior art foamed polymer composition (Figure 5A) and a foamed polymer blend of the present invention (Figure 5B).

Figures 6A and 6B show electron micrographs of a prior art foamed polymer composition (Figure 6A) and a foamed polymer blend of the present invention (Figure 6B).

Figures 7A and 7B show die swell data for prior art polymer blends (Figure 7B) and several polymer blends used for the foams of the present invention (Figure 7A).

DETAILED DESCRIPTION OF THE INVENTION

In one embodiment, the present invention provides a foamed polymer composition formed of a blend of a first polymer component and a second polymer component. The blended components are foamed using conventional

chemical or physical foaming or blowing agents to form the unique and advantageous foamed polymer blends of the present invention. Each of the components of the invention is described in detail below.

5.1. First Polymer Component

The first polymer component is a thermoplastic material based on polypropylene and having substantial crystallinity. The first polymer component, or "FPC", can be a polypropylene homopolymer, a random copolymer of propylene and an olefinic comonomer, or a block or graft copolymer of propylene and an olefinic comonomer. The first polymer component can also be a blend of two or more propylene-based polymers or copolymers.

When the first polymer component is or includes a polypropylene homopolymer, the polypropylene is preferably substantially isotactic polypropylene. The degree of isotacticity required can be characterized by the polymer melting point; *i.e.*, the polypropylene should have sufficient stereoregularity such that the polymer has a melting point of greater than 110°C, preferably greater than 115°C, and more preferably greater than 130°C.

When the first polymer component is a propylene copolymer, the copolymer can include less than 10% by weight of comonomer, preferably less than 8% by weight, and more preferably less than 5% by weight. Smaller amounts of comonomer can also be used, such as less than 4%, less than 3%, or less than 2% by weight. The propylene units of the copolymer should be substantially isotactic, such that the copolymer has a melting point of greater than 110°C, preferably greater than 115°C, and more preferably greater than 130°C.

Suitable comonomers include ethylene and α -olefins, such as C₄-C₂₀ α -olefins and preferably C₄-C₁₂ α -olefins. The α -olefin comonomer can be linear or branched, and two or more comonomers can be used, if desired. Examples of suitable comonomers include ethylene, linear C₄-C₁₂ α -olefins, and α -olefins having one or more C₁-C₃ alkyl branches. Specific examples include ethylene; 3-methyl-1-butene; 3,3-dimethyl-1-butene; 1-pentene; 1-pentene with one or more methyl, ethyl or propyl substituents; 1-hexene with one or more methyl, ethyl or propyl substituents; 1-heptene with one or more methyl, ethyl or propyl

substituents; 1-octene with one or more methyl, ethyl or propyl substituents; 1-nonene with one or more methyl, ethyl or propyl substituents; ethyl, methyl or dimethyl-substituted 1-decene, or 1-dodecene. Preferred comonomers include ethylene, 1-butene, 1-pentene, 3-methyl-1-butene, 1-hexene, 3-methyl-1-pentene, 4-methyl-1-pentene, 3,3-dimethyl-1-butene, 1-heptene, 1-hexene with a methyl substituent on any of C₃-C₅, 1-pentene with two methyl substituents in any stoichiometrically acceptable combination on C₃ or C₄, 3-ethyl-1-pentene, 1-octene, 1-pentene with a methyl substituent on any of C₃ or C₄, 1-hexene with two methyl substituents in any stoichiometrically acceptable combination on C₃-C₅, 1-pentene with three methyl substituents in any stoichiometrically acceptable combination on C₃ or C₄, 1-hexene with an ethyl substituent on C₃ or C₄, 1-pentene with an ethyl substituent on C₃ and a methyl substituent in a stoichiometrically acceptable position on C₃ or C₄, 1-decene, 1-nonene, 1-nonene with a methyl substituent on any of C₃-C₉, 1-octene with two methyl substituents in any stoichiometrically acceptable combination on C₃-C₇, 1-heptene with three methyl substituents in any stoichiometrically acceptable combination on C₃-C₆, 1-octene with an ethyl substituent on any of C₃-C₇, 1-hexene with two ethyl substituents in any stoichiometrically acceptable combination on C₃ or C₄, and 1-dodecene. It should be appreciated that the list of comonomers above is merely exemplary, and is not intended to be limiting. A particularly preferred comonomer is ethylene.

The first polymer component can be prepared by any method suitable for producing polypropylene or polypropylene-based copolymers having the properties described above. Such methods are well-known in the art, and can include single stage or multiple stage reactor processes carried out under high pressure, slurry, gas, bulk or solution phase conditions, or a combination thereof. Suitable catalysts include traditional Ziegler-Natta catalysts or single-site metallocene catalyst systems. The catalyst used is preferably one which has a high degree of isospecificity. Polymerization can be carried out by a continuous or batch process, and may include the use of chain transfer agents, scavengers or other additives that are deemed suitable. One skilled in the art can readily determine the appropriate conditions, reagents and catalysts to produce the first

polymer component as described above.

5.2 Second Polymer Component

The second polymer component, or "SPC", includes at least one
5 copolymer of propylene and at least one comonomer, the comonomer being
ethylene or an α -olefin. Preferred polypropylene copolymers of this
embodiment can be prepared by polymerizing propylene and ethylene or a C_4 - C_{30}
 α -olefin, most preferably propylene and ethylene, in the presence of a chiral
metallocene catalyst with an activator and optional scavenger. In this
10 embodiment, preferred comonomers include ethylene and linear or branched C_4 to
 C_{30} α -olefins, or combinations thereof. Preferred linear α -olefins include ethylene
and C_4 to C_8 α -olefins, more preferably ethylene, 1-butene, 1-hexene, and
1-octene, even more preferably ethylene or 1-butene. Preferred branched
 α -olefins include 4-methyl-1-pentene, 3-methyl-1-pentene, and
15 3,5,5-trimethyl-1-hexene. The propylene copolymer of the SPC is preferably a
random copolymer, as the term is defined herein below.

The polypropylene copolymer of the SPC is semi-crystalline; *i.e.*, the
polypropylene copolymer has a crystallinity of from 2% to 65%. Within this
range of crystallinity, alternative lower limits of crystallinity can be 5% or 10%,
20 and alternative upper limits of crystallinity can be 50%, 45% or 40%, with ranges
from any lower limit to any upper limit being included within the scope of the
invention. These measurements of the crystallinity are by Differential Scanning
Calorimetry with reference to the heat of fusion of pure homoisotactic
polypropylene by the Huffman-Weeks extrapolation procedure to be 189J/g.

The crystallinity of the polypropylene copolymer of the SPC is derived
25 from isotactic polypropylene sequences in the copolymer. The amount of
propylene in the SPC can be from 65% to 95% by weight. Within this range,
alternative lower limits of propylene content in the SPC can be 70% or 80% by
weight, and alternative upper limits of propylene content can be 92.5%, 90%, or
30 89% by weight, with ranges from any lower limit to any upper limit being
included within the scope of the invention. The balance of the copolymer is one

or more α -olefins as described above and optionally minor amounts of one or more diene comonomers.

The semi-crystalline polypropylene copolymer necessarily has a non-zero heat of fusion, due to the measurable crystallinity. The crystallinity can be calculated from the heat of fusion, using a preferred value of 189 J/g for 100% crystallinity and a linear relationship between heat of fusion and crystallinity; see, B. Wunderlich, "Macromolecular Physics," vol. 3, Academic Press (1980), esp. Chapter 8.4.2.

The polypropylene copolymer of the SPC preferably has a single broad melting transition. Typically, a sample of the polypropylene copolymer will show secondary melting peaks or shoulders adjacent to the principal peak, and this combination is considered together as single melting point, *i.e.*, a single broad melting transition. The highest of these peaks is considered the melting point. The polypropylene copolymer preferably has a melting point of from 25°C to 110°C. Within this range, alternative lower limits of the melting point can be 30°C or 35°C, and alternative upper limits of the melting point can be 105°C or 90°C, with ranges from any lower limit to any upper limit being included within the scope of the invention.

The weight average molecular weight of the polypropylene copolymer can be from 10,000 to 5,000,000 g/mol, preferably 80,000 to 500,000 with a MWD (M_w/M_n) from 1.5 to 40, more preferably from 1.8 to 5 and most preferably from 1.8 to 3. In another embodiment, it is preferred that the polypropylene copolymer has a ML (1+4)@125°C less than 100, more preferably less than 75, even more preferably less than 60, most preferably less than 30.

The polypropylene copolymer of the present invention preferably is a random, crystallizable copolymer having a narrow compositional distribution. The intermolecular composition distribution of the polymer is determined by thermal fractionation in a solvent. A typical solvent is a saturated hydrocarbon such as hexane or heptane. The thermal fractionation procedure is described below. Typically, approximately 75% by weight and more preferably 85% by weight of the polymer is isolated as one or two adjacent, soluble fractions, with the balance of the polymer in immediately preceding or succeeding fractions.

Each of these fractions has a composition (wt. % ethylene content) with a difference of no greater than 20% (relative) and more preferably no greater than 10% (relative) from the average weight % ethylene content of the polypropylene copolymer. For purposes of the present disclosure, the polypropylene copolymer is considered to have a "narrow" compositional distribution if it meets the fractionation test outlined above.

The length and distribution of stereoregular propylene sequences in preferred polypropylene copolymers is consistent with substantially random statistical copolymerization. It is well known that sequence length and distribution are related to the copolymerization reactivity ratios. As used herein, the term "substantially random" means a copolymer for which the product of the reactivity ratios is generally 2 or less. In contrast, in stereoblock structures, the average length of PP sequences is greater than that of substantially random copolymers with a similar composition. Prior art polymers with stereoblock structure have a distribution of PP sequences consistent with these "blocky" structures rather than a random, substantially statistical distribution.

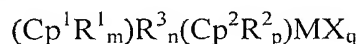
The reactivity ratios and sequence distribution of the polymer may be determined by C-13 NMR, which locates the ethylene residues in relation to the neighboring propylene residues. To produce a crystallizable copolymer with the required randomness and narrow composition distribution, it is desirable to use: (1) a single-sited catalyst; and (2) a well-mixed, continuous flow, stirred tank polymerization reactor which allows only a single polymerization environment for substantially all of the polymer chains of preferred polypropylene copolymers.

Preferred polypropylene copolymers of this embodiment are prepared by polymerizing propylene and at least one comonomer selected from the group consisting of ethylene and C₄-C₂₀ alpha-olefins, preferably ethylene, in the presence of a chiral metallocene catalyst, with an activator and optional scavenger. Preferred chiral metallocenes are those known to favor: (1) incorporation of propylene to form predominantly isotactic polypropylene pentads; and (2) statistically random incorporation of the α -olefin comonomer(s).

The terms "metallocene" and "metallocene catalyst precursor" are terms known in the art to mean compounds having a Group 4, 5 or 6 transition metal

(M), with a cyclopentadienyl (Cp) ligand or ligands which may be substituted, at least one non-cyclopentadienyl-derived ligand (X), and zero or one heteroatom-containing ligand (Y), the ligands being coordinated to M and corresponding in number to the valence thereof. The metallocene catalyst precursors generally require activation with a suitable co-catalyst (referred to as an "activator"), in order to yield an "active metallocene catalyst", *i.e.*, an organometallic complex with a vacant coordination site that can coordinate, insert, and polymerize olefins.

Preferred metallocenes are cyclopentadienyl complexes which have two Cp ring systems as ligands. The Cp ligands preferably form a bent sandwich complex with the metal, and are preferably locked into a rigid configuration through a bridging group. These cyclopentadienyl complexes are preferably chiral and have the general formula:



wherein Cp^1 and Cp^2 are preferably the same; R^1 and R^2 are each, independently, a halogen or a hydrocarbyl, halocarbyl, hydrocarbyl-substituted organometalloid or halocarbyl-substituted organometalloid group containing up to 20 carbon atoms; m is preferably 1 to 5; p is preferably 1 to 5; preferably two R^1 and/or R^2 substituents on adjacent carbon atoms of the cyclopentadienyl ring associated therewith can be joined together to form a ring containing from 4 to 20 carbon atoms; R^3 is a bridging group; n is the number of atoms in the direct chain between the two ligands and is preferably 1 to 8, most preferably 1 to 3; M is a transition metal having a valence of from 3 to 6, preferably from group 4, 5, or 6 of the periodic table of the elements, and is preferably in its highest oxidation state; each X is a non-cyclopentadienyl ligand and is, independently, a hydrocarbyl, oxyhydrocarbyl, halocarbyl, hydrocarbyl-substituted organometalloid, oxyhydrocarbyl-substituted organometalloid or halocarbyl-substituted organometalloid group containing up to 20 carbon atoms; and q is equal to the valence of M minus 2.

Numerous examples of the biscyclopentadienyl metallocenes described above for the invention are disclosed in U.S. Patent Nos. 5,324,800; 5,198,401; 5,278,119; 5,387,568; 5,120,867; 5,017,714; 4,871,705; 4,542,199; 4,752,597;

5,132,262; 5,391,629; 5,243,001; 5,278,264; 5,296,434; and 5,304,614, all of which are incorporated herein by reference for purposes of U.S. patent practice.

Illustrative, but not limiting examples of preferred biscyclopentadienyl metallocenes of the type described above are the racemic isomers of:

5 $\mu\text{-(CH}_3)_2\text{Si(indenyl)}_2\text{M(Cl)}_2$,
 $\mu\text{-(CH}_3)_2\text{Si(indenyl)}_2\text{M(CH}_3)_2$,
 $\mu\text{-(CH}_3)_2\text{Si(tetrahydroindenyl)}_2\text{M(Cl)}_2$,
 $\mu\text{-(CH}_3)_2\text{Si(tetrahydroindenyl)}_2\text{M(CH}_3)_2$,
 $\mu\text{-(CH}_3)_2\text{Si(indenyl)}_2\text{M(CH}_2\text{CH}_3)_2$, and
10 $\mu\text{-(C}_6\text{H}_5)_2\text{C(indenyl)}_2\text{M(CH}_3)_2$,
wherein M is Zr, Hf, or Ti.

A preferred polypropylene copolymer used in the present invention is described in detail as the "Second Polymer Component (SPC)" in co-pending U.S. Application Nos. 09/569,362, filed May 11, 1999 and 09/342,854, filed June 29,
15 1999, and described in further detail as the "Propylene Olefin Copolymer" in U.S. Application No. 09/346,460, filed July 2, 1999, which are all fully incorporated by reference herein for purposes of U.S. practice.

In addition to one or more comonomers (*i.e.*, α -olefins) such as, but not limited to, ethylene and α -olefins having 4 to 8 carbon atoms, and styrenes, the
20 polypropylene copolymers as described above can optionally contain long chain branches. These can optionally be generated using one or more α,ω -dienes.

In another embodiment, the SPC can be a blend of discrete polymers. Such blends can be of two or more polyethylene copolymers (as described above), two or more polypropylene copolymers (as described above), or at least one of
25 each such polyethylene copolymer and polypropylene copolymer, where each of the components of the SPC blend would individually meet the criteria of an SPC as described herein.

It should be understood in the context of the present invention that, in one embodiment, more than one second polymer component may be used in a single
30 blend with a first polymer component. Each second polymer component can be as described above, and the number of second polymer components in this

embodiment is three or more preferably, two. In this embodiment, the second polymer components differ in their alpha-olefin content, with one being in the range of 5 wt % to 9 wt% alpha-olefin, while the other is in the range of 10 wt% to 22 wt% alpha-olefin. The preferred alpha-olefin is ethylene. It is believed that the use of two second polymer components in conjunction with a single first polymer component leads to beneficial improvements in the tensile-elongation properties of the blends.

5.3 Blend of First and Second Polymer Components

The first polymer component and the second polymer component are blended together to form a foamable polymer composition. The blended composition includes from 50% to 99.9% by weight of the first polymer component, based on the total weight of the first and second polymer components. Within this range of weight percents, alternative lower limits of the weight percent of the first polymer component can be 70% or 80%, and alternative upper limits of weight percent of the first polymer component can be 99.5% or 99%, with ranges from any lower limit to any upper limit being included within the scope of the invention.

The blends of first polymer component and second polymer component of the present invention may be prepared by any procedure that provides an intimate admixture of the components. As a specific, non-limiting example, the components can be combined by melt pressing the components together on a Carver press to a thickness of 0.5 mm (20 mils) and a temperature of 180°C, rolling up the resulting slab, folding the ends together, and repeating the pressing, rolling, and folding operation 10 times. Internal mixers are particularly useful for solution or melt blending. Blending at a temperature of 180°C to 240°C in a Brabender Plastograph for 1 to 20 minutes has been found satisfactory. Still another method that may be used for admixing the components involves blending the polymers in a Banbury internal mixer above the flux temperature of all of the components, *e.g.*, 180°C for 5 minutes. The complete admixture of the polymeric components is indicated by the narrowing of the crystallization and melting transitions characteristic of the polypropylene crystallinity of the components to

give a single or a small range of crystallization and melting points for the blend. These batch-mixing procedures are typically supplanted by continuous mixing processes in the industry. These processes are well known in the art and include single and twin screw mixing extruders, static mixers for mixing molten polymer streams of low viscosity, impingement mixers, as well as other machines and processes designed to disperse the first polymer component and the second polymer component in intimate contact.

5.4 Foamed Compositions

The terms "foamed polymer composition" or "polymer foam" are used interchangeably herein to mean a system having a gas phase and at least one solid phase, where the solid phase is continuous and formed of a polymer composition, and the gas phase is distributed throughout the solid phase as a plurality of voids, also referred to as pores, pockets or cells. The cells can be open (interconnected) or closed (isolated). Other terms commonly used in the literature, and intended to be within the scope of the terms "foamed polymer composition" and "polymer foam" are "cellular plastic", "foamed plastic", "expanded plastic" and "plastic foam".

The blend of first polymer component and second polymer component can be foamed by any of a variety of chemical or physical foaming processes known in the art. As used herein, the terms "foaming agent" and "blowing agent" are used interchangeably, and are intended to include both physical and chemical agents unless specified otherwise. Physical foaming or blowing agents include various gases which are chemically unreactive toward the polymer blend. Physical blowing agents can be mechanically whipped or frothed into the polymer blend to form a foam, or dispersed or dissolved in the blend, possibly at elevated pressure, and subsequently liberated at reduced pressure and/or by heating, thereby foaming the blend.

Depending upon the blowing agent used, the foamed polymer compositions of the present invention can have densities of 0.5 g/cm^3 (500 kg/m^3) or less, and can be as low as 0.1 g/cm^3 (300 kg/m^3) or 0.3 g/cm^3 (400 kg/m^3), or densities greater than 0.5 g/cm^3 (500 kg/m^3) and as high as 0.6 g/cm^3 (600 kg/m^3)

or 0.7 g/cm³ (700 kg/m³). In general, physical blowing agents are typically used when low foam densities (*e.g.*, 0.5 g/cm³ or less) are desired, and chemical blowing agents are typically used for higher densities.

Foam density can be calculated by measuring the physical dimensions of the foam sample and taking the ratio of mass to volume. In addition, a crude estimate of density can be made by a floatation technique. Hardness of the foam can be measured following the procedure of ASTM D 2240-86; the foams of the present invention have hardnesses of 45-60 Shore A. Foam cell size can be measured by measuring the size of cell cross-sections optically, such as by optical microscope or scanning electron microscopy (SEM). Foams of the present invention typically have cell sizes of 20 to 1000 microns. Other parameters useful to characterize these foams include a compression set of 40-80% recovery after 30 min. (ISO 1856); a compression modulus of 800 to 2000 psi (ISO 844); and a compressive stress at 10% strain of 5-20 pounds (ASTM B1621).

Examples of physical blowing agents include non-reactive inorganic gases or gas mixtures, such as air, carbon dioxide, nitrogen, helium and argon; hydrocarbon gases or low-boiling liquids, such as butane, isobutane, pentane, neopentane, hexane, isohexanes, isoheptanes, benzene and toluene; halogenated hydrocarbons, such as methyl chloride, dichloroethane, trichloroethylene and methylene chloride; chlorofluorocarbons (CFCs, or Freons), such as trichlorofluoromethane (CCl₃F, Freon 11), dichlorodifluoromethane (CCl₂F₂, Freon 12), trifluorochloromethane (CF₃Cl, Freon 13), 1,2,2-trichlorotrifluoroethane (CFCl₂-CF₂Cl, Freon 113) and 1,2-dichlorotetrafluoroethane (CClF₂-CClF₂, Freon 114); and Freon alternatives, such as various HCFCs. Because of concerns over the health and environmental effects of hydrocarbons, halogenated hydrocarbons and CFCs, the non-reactive inorganic gases or gas mixtures are preferred physical blowing agents.

Chemical blowing agents are compounds or mixtures which liberate gases such as nitrogen, carbon dioxide or ammonia by thermal decomposition or chemical reaction. Examples of chemical blowing agents include ammonium carbonate; alkali metal carbonates and bicarbonates; azo and diazo compounds, such as azodinitriles (azodiisobutyronitrile, azodicyclohexyldicyanide),

2,2'-dialkoxy-2,2'-azopropanes, azodicarboxylic acid derivatives (azodicarboxamide, N,N'-di(t-butyl)azodicarboxamide, polyazocarboxamides, azodicarboxylic acid esters and salts) and diazoaminobenzene (DAB); N-nitroso compounds, such as N,N'-dinitrosopentamethylenetetramine (DNPA) and
5 N,N'-dimethyl-N,N'-dinitrosoterephthalamide (DTA); sulfonylhydrazides, such as benzosulfonylhydrazide (BSH), p-toluenesulfonylhydrazide, 4,4'-oxybis(benzenesulfonylhydrazide) (OBSH), bis(hydrazosulfonyl)benzene, 4,4'-bis(hydrazosulfonyl)biphenyl and diphenylsulfone-3,3'-disulfonylhydrazide; carbazides, such as terephthalazide; sulfazides, such as
10 p-methoxycarbonylaminobenzenesulfide, 4,4'-bis(sulfazido)diphenyloxide, benzenesulfazide, p-chlorobenzenesulfazide, p-toluenesulfazide, benzene-1,3-disulfazide and azidopentaphenylphosphonitrile; triazine derivatives, such as 2,4,6-trihydrazino-1,3,5-triazine and 2,4,6-sulfonylhydrazo-S-triazines; tri- and tetrazole derivatives, such as 5(N-morpholinyl)-1-thia-2,3,4-triazole,
15 N-hydroxybenzotriazole, 5-amino-4-mercapto-1,2,3-(2H)triazole, 5-hydroxytetrazole and ammonium 5-azidotetrazole; sulfonyl semicarbazides, such as p-toluenesulfonyl semicarbazide, methyl sulfonyl semicarbazide and 4,4'-oxybis(benzosulfonyl semicarbazide); urea derivatives, such as urea oxalate, nitrourea, methylaminonitrourea and N,N'-dinitrosoethylurea; guanidine
20 derivatives, such as aminoguanidine carbonate; mixtures of acids and metals; mixtures of organic acids with inorganic carbonates; and mixtures of nitrites and ammonium salts.

When used at low levels, generally less than 1% by weight, and typically 0.25% by weight, based on the weight of the polymer blend, chemical blowing
25 agents may function as bubble nucleating agents, and facilitate the formation of more uniformly-sized bubbles. This nucleating function is often utilized even when the primary foaming agent is a physical blowing agent, such as carbon dioxide gas. Talc can also be used for bubble nucleation, if desired.

It should be appreciated that the physical and chemical blowing agents
30 listed above are merely exemplary, and not limiting, and some may be better suited than others for particular polymer blends, foam characteristics, or process conditions. One skilled in the art can readily choose a suitable blowing agent

based on these and other criteria well-known in the art. Further examples of blowing agents and foaming methods can be found in Kirk-Othmer, *Encyclopedia of Chemical Technology*, Fourth Edition, Vol. 11, p.730-783, and in Klemptner and Frisch (eds.), *Handbook of Polymeric Foams and Foam Technology*, Hanser Publishers (1991), the disclosures of which are incorporated herein by reference, for purposes of U.S. patent practice.

It should be appreciated that while the above discussion has been focussed on a foamed polymer composition, the invention in another embodiment is directed to an article formed of, or including, the polymer foams of the present invention.

5.5 Additional Features

The polymer blends of the instant invention exhibit a remarkable combination of desirable physical properties. The incorporation of as little as 5% first polymer component in the blend copolymers increases the propylene sequence melting point or the polymer softening point, but more significantly, reduces the range as compared to the SPC. In addition, the incorporation of a first polymer component in accordance with the instant invention nearly eliminates the stickiness characteristic of the SPC. Further, the thermal characteristics of the copolymer blends are markedly improved over those of the second polymer component.

The crystallization temperature and the melting point of the blends are changed as a result of the blending operation. In an embodiment of the invention, the blend of first polymer component and second polymer component has single crystallization temperature and melting point. These temperatures are higher than the corresponding temperatures for the second polymer component and close to that of the first polymer component. In other embodiments, the second polymer component and the first polymer component have distinct melting and crystallization temperatures but have these temperatures closer together than would be expected for a combination of the second polymer component and the first polymer component. In all these cases, the glass transition temperature of the second polymer component is retained in the polymer blend.

This favorable combination of thermal properties, together with good processing properties, permits the satisfactory use of the polymer blends of the present invention in foam operations without premature collapse of the foam profile that is typically encountered with the FPC alone. Foamed articles prepared from the instant copolymer blends accordingly exhibit excellent long-term dimensional stability. The advantages referred to above are obtained without the need for elaborate purification of the polypropylene copolymer of the second polymer component, or the tedious preparation of a carefully structured block copolymer. Further, by the use of the second polymer component and the first polymer component, a blend can be obtained with a lower glass transition temperature than would be expected for a random copolymer of the same composition as the blend. In particular, the glass transition temperature of the blend is closer to that of the second polymer component and lower than the glass transition temperature of the first polymer component. This can be accomplished without an exceptionally high alpha-olefin content in the polymer blend which we believe, while not meant to be limited thereby, would lead to degradation of the tensile-elongation properties of the blend.

The mechanism by which the desirable characteristics of the present copolymer blends are obtained is not fully understood. However, it is believed to involve a co-crystallization phenomenon between propylene sequences of similar stereoregularity in the various polymeric components, which results, in one embodiment, in a single crystallization temperature and a single melting temperature of the copolymer blend which is higher than those of the second polymer component alone. In another embodiment, the combination of the first polymer component and the second polymer component has a melting point closer together than would be expected on a comparison of the properties of the individual components alone. It is surprising that in the one embodiment, the blend has a single crystallization temperature and a single melting temperature, since it would be expected by those skilled in the art that the blending of two crystalline polymers would result in a double crystallization temperature as well as a double melting temperature reflecting the two polymeric components. However, the intimate blending of the polymers having the required crystallinity

characteristics apparently results in a crystallization phenomenon that modifies the other physical properties of the second polymer component, thus measurably increasing its commercial utility and range of applications.

While the above discussion has been limited to the description of the invention in relation to having only the first and second polymer components, as will be evident to those skilled in the art, the polymer blend compositions of the present invention may include other components, such as additives which may be present in the composition to enhance a specific property, or as a result of processing of the individual components. Additives which may be incorporated include, for example, fire retardants, antioxidants, plasticizers, and pigments. Other additives which may be used to enhance properties include antiblocking agents, coloring agents, stabilizers, and oxidative-, thermal-, and ultraviolet-light-inhibitors. Lubricants, mold release agents, nucleating agents, reinforcers, and fillers (including granular, fibrous, or powder-like fillers) may also be used. Nucleating agents and fillers tend to improve rigidity of the article, and are preferred when increased rigidity is desired.

The list of additives described above is not intended to be inclusive of all types of additives which may be used with the present invention. Those skilled in the art will appreciate that other additives may be used to enhance various properties of the composition, and the polymer blend compositions of the present invention may be modified to adjust the characteristics of the blend, and thus resistant foam quality, as desired.

EXAMPLES

Methods and Materials

Mooney viscosity of various samples was measured as ML (1+4) at 125°C in Mooney units according to ASTM D-1646.

The composition of ethylene-propylene copolymers used as comparative examples below was measured as weight percent ethylene according to ASTM D-3900.

The composition of the second polymer component was measured as weight percent according to the following technique. A thin homogeneous film of

the second polymer component, pressed at a temperature of 150°C or greater, was mounted on a Perkin Elmer PE 1760 infrared spectrophotometer. A full spectrum of the sample from 600 cm⁻¹ to 4000 cm⁻¹ was recorded, and the ethylene weight percent of the second polymer component was calculated according to the following calibration curve:

$$\text{ethylene wt \%} = 82.585 - 111.987 X + 30.045 X^2$$

where X is the ratio of the peak height at 1155 cm⁻¹ to the peak height at either 722 cm⁻¹ or 732 cm⁻¹, whichever is higher.

Techniques for determining the molecular weight (Mn and Mw) and molecular weight distribution (MWD) can be found in U.S. Patent No. 4,540,753 to Cozewith et al. and references cited therein (the disclosures of which are incorporated herein by reference for purposes of U.S. practice), and in Verstrate et al., *Macromolecules*, vol. 21, p.3360 (1988) and references cited therein (the disclosures of which are incorporated herein by reference for purposes of U.S. practice).

Thermodynamic heat of fusion data were determined by differential scanning calorimetry (DSC), the procedure for which is as follows. 6 to 10 mg of a sheet of the polymer pressed at approximately 200°C to 230°C is removed with a punch die. This sample is annealed at room temperature for 80 to 100 hours. At the end of the annealing period, the sample is placed in a differential scanning calorimeter (Perkin Elmer 7 Series Thermal Analysis System) and cooled to -50°C to -70°C. The sample is then heated at a rate of 20°C/min to a final temperature of 200°C to 220°C. The thermal output is recorded as the area under the melting peak curve of the sample, which is typically peaked at 30°C to 175°C, and occurs between the temperatures of 0°C and 200°C. The thermal output in joules is a measure of the heat of fusion. The melting point is recorded as the temperature of the greatest heat absorption within the range of melting of the sample. Under these conditions, the melting point of the second polymer component and its heat of fusion are lower than the corresponding values of the first polymer component as outlined in the description above.

The composition distribution of the second polymer component was measured as described below. 30 g of the second polymer component is cut into

small cubes measuring 1/8" (0.3 cm) on each side. The cubes are introduced into a thick-walled glass bottle, along with 50 mg of Irganox 1076, an antioxidant commercially available from Ciba-Geigy Corporation. Then, 425 mL of hexane (a principal mixture of normal and iso isomers) is added to the contents of the bottle, the bottle is closed with a screw cap, and the sealed bottle is maintained at 23°C for 24 hours. At the end of this period, the solution is decanted and the residue is treated with additional hexane for an additional 24 hours. At the end of this period, the two hexane solutions are combined and evaporated to yield a residue of the polymer soluble at 23°C. To the residue is added sufficient hexane to bring the volume to 425 mL, and the bottle is maintained at 31°C for 24 hours in a covered circulating water bath. The soluble polymer is decanted, and an additional amount of hexane is added for another 24 hours at 31°C prior to decanting again. In this same manner, fractions of the second polymer component soluble at 40°C, 48°C, 55°C and 62°C are obtained at temperature intervals of approximately 8°C between stages. Further increases in temperature to 95°C can be accommodated, if heptane, instead of hexane, is used as the solvent for all temperatures above 60°C. The soluble polymers are dried, weighed and analyzed for composition, as weight percent ethylene content, by the IR technique described above. Soluble fractions obtained in the adjacent temperature regions are the adjacent fractions in the specification above.

The elongation is determined for blends at 20in/min according to the procedure described in ASTM D790. The data is reported in engineering units with no correction to the stress for the lateral contraction in the specimen due to tensile elongation. The stress-strain elongation properties of the insitu and the corresponding physical blends was evaluated using dumbbell shaped sample. The samples were compression molded at 180° C to 200° C for 15 minutes at a force of 15 tons into a plaque of dimensions of 6 in x 6 in. The cooled plaques were removed and the specimens were removed with a die. The stress strain evaluation of the samples was conducted on an Instron 4465, made by Instron Corporation of 100 Royall Street, Canton, MA. The digital data was collected in a file collected by the Series IX Material Testing System available from Instron Corporation and

analyzed using Excel 5, a spreadsheet program available from Microsoft Corporation of Redmond, WA.

Flexural modulus (secant 1%) was determined according to according to ASTM D790.

5 Die swell, i.e., the ratio of extruded polymer diameter to die diameter, was determined as follows. The polymer was heated to the softening temperature (typically 190°C to 230°C) and driven by a piston through a die orifice of predetermined length (L), diameter (D) and L/D ratio. The extruded polymer was allowed to cool and solidify, then the diameter of the extruded polymer was
10 measured using a micrometer. This procedure is consistent with ASTM D-3825-96 and D-1238.

EPR in the data tables below is Vistalon® 457, an EP rubber sold by the ExxonMobil Chemical Company, Houston TX.

The invention, while not meant to be limited thereby, is further illustrated
15 by the following specific examples.

Example 1: Ethylene-Propylene Copolymerization to Form the Second
Polymer Component

Polymerizations were conducted in a 1-liter thermostatted continuous feed
20 stirred tank reactor using hexane as the solvent. The polymerization reactor was full of liquid. The residence time in the reactor was typically 7-9 minutes and the pressure was maintained at 400 kPa. Hexane, ethene and propene were metered into a single stream and cooled before introduction into the bottom of the reactor. Solutions of all reactants and polymerization catalysts were introduced
25 continuously into the reactor to initiate the exothermic polymerization. Temperature of the reactor was maintained at 41°C by changing the temperature of the hexane feed and by circulating water in an external jacket. For a typical polymerization, the temperature of the feed was 0°C.

Ethene was introduced at the rate of 45 g/min and propene was introduced
30 at the rate of 480 g/min. The polymerization catalyst, dimethyl silyl bridged bis-indenyl hafnium dimethyl activated 1:1 molar ratio with N',N'-dimethyl anilinium-tetrakis(pentafluorophenyl)borate was introduced at the rate of 0.00897

g/hr. A dilute solution of triisobutyl aluminum was introduced into the reactor as a scavenger of catalyst terminators; a rate of approximately 28.48 mol of scavenger per mol of catalyst was adequate for this polymerization. After five residence times of steady polymerization, a representative sample of the polymer produced in this polymerization was collected. The solution of the polymer was withdrawn from the top, and then steam distilled to isolate the polymer. The rate of formation of the polymer was 285.6 g/hr. The polymer produced in this polymerization had an ethylene content of 13%, ML@125 (1+4) of 12.1, and had isotactic propylene sequences.

Variations in the composition of the polymer were obtained principally by changing the ratio of ethene to propene. Molecular weight of the polymer could be increased by a greater amount of ethene and propene compared to the amount of the polymerization catalyst. Dienes such as norbornene and vinyl norbornene could be incorporated into the polymer by adding them continuously during polymerization.

Example 2: Comparative Ethylene-Propylene Polymerization Where the Propylene Residues are Atactic

Polymerizations were conducted in a 1-liter thermostatted continuous feed stirred tank reactor using hexane as the solvent. The polymerization reactor was full of liquid. The residence time in the reactor was typically 7-9 minutes and the pressure was maintained at 400 kPa. Hexane, ethene and propene were metered into a single stream and cooled before introduction into the bottom of the reactor. Solutions of all reactants and polymerization catalysts were introduced continuously into the reactor to initiate the exothermic polymerization. Temperature of the reactor was maintained at 45°C by changing the temperature of the hexane feed and by using cooling water in the external reactor jacket. For a typical polymerization, the temperature of the feed was -10°C. Ethene was introduced at the rate of 45 g/min and propene was introduced at the rate of 310 g/min. The polymerization catalyst, dimethyl silyl bridged (tetramethylcyclopentadienyl) cyclododecylamido titanium dimethyl activated 1:1 molar ratio with N',N'-dimethyl anilinium-tetrakis(pentafluorophenyl)borate was

introduced at the rate of 0.002780 g/hr. A dilute solution of triisobutyl aluminum was introduced into the reactor as a scavenger of catalyst terminators; a rate of approximately 36.8 mole per mole of catalyst was adequate for this polymerization. After five residence times of steady polymerization, a representative sample of the polymer produced in this polymerization was collected. The solution of the polymer was withdrawn from the top, and then steam distilled to isolate the polymer. The rate of formation of the polymer was 258 g/hr. The polymer produced in this polymerization had an ethylene content of 14.1 wt%, and ML@125 (1+4) of 95.4.

Variations in the composition of the polymer were obtained principally by changing the ratio of ethene to propene. Molecular weight of the polymer could be increased by a greater amount of ethene and propene compared to the amount of the polymerization catalyst. These polymers are described as "aePP" in the Tables herein.

Example 3: Analysis and Solubility of Second Polymer Components

In the manner described in Example 1, several second polymer components (labeled SPC-1 to SPC-5) were synthesized. Various properties of these polymers were measured as described above. The results are shown in Tables 1-3, along with EPR and aePP for comparison.

Table 1 shows the molecular weights, ethylene content, melting points, heats of fusion and Mooney viscosities of the samples.

Table 1

| Sample | Mn (g/mol) | Mw (g/mol) | Ethylene (wt %) | ΔH_{fusion} (J/g) | Melting Point (°C) | ML (1+4)@125°C |
|---------------------|---------------|---------------|--------------------|-------------------------------------|-----------------------|-------------------|
| SPC-1 | 102,000 | 248,900 | 7.3 | 71.9 | 84.7 | 14 |
| SPC-2 | 124,700 | 265,900 | 11.6 | 17.1 | 43.0 | 23.9 |
| SPC-3 | 121,900 | 218,900 | 16.4 | 7.8 | 42.2 | 33.1 |
| SPC-4 | -- | -- | 11.1 | 25.7 | 63.4 | 34.5 |
| SPC-5 | -- | -- | 14.7 | 13.2 | 47.8 | 38.4 |
| EPR ^(a) | -- | -- | 47.8 | nd ^(b) | nd ^(b) | 40 |
| aePP ^(a) | -- | -- | 11.7 | nd ^(b) | nd ^(b) | 23 |

^(a) comparative example^(b) not detected

Table 2 shows the solubility of the second polymer component and the comparative samples in hexane at several temperatures.

Table 2

| Sample | Amount Soluble at T (weight %) | | | |
|---------------------|--------------------------------|------|------|------|
| | 23°C | 31°C | 40°C | 48°C |
| SPC-1 | 1.0 | 2.9 | 28.3 | 68.5 |
| SPC-2 | 6.5 | 95.7 | -- | -- |
| SPC-3 | 51.6 | 52.3 | 2.6 | -- |
| SPC-4 | 18.7 | 83.6 | -- | -- |
| SPC-5 | 36.5 | 64.2 | -- | -- |
| EPR ^(a) | 101.7 | -- | -- | -- |
| aePP ^(a) | 100.5 | -- | -- | -- |

^(a) values may add to more than 100% due to imperfect sample drying^(b) comparative example

Table 3 shows the composition of the fractions of the second polymer components of Table 2. Only fractions greater than 4% of the total polymer mass were analyzed. The values in Table 3 are believed to be accurate to ± 0.4 wt%

absolute.

Table 3

| Sample | Weight % Ethylene in Fraction Soluble at T | | | |
|---------------------|--|------|------|------|
| | 23°C | 31°C | 40°C | 48°C |
| SPC-1 | -- | -- | 8.0 | 7.6 |
| SPC-2 | 12.0 | 11.2 | -- | -- |
| SPC-3 | 16.8 | 16.5 | -- | -- |
| SPC-4 | 13.2 | 11.2 | -- | -- |
| SPC-5 | 14.9 | 14.6 | -- | -- |
| EPR ^(a) | 46.8 | -- | -- | -- |
| aePP ^(a) | 11.8 | -- | -- | -- |

^(a) comparative example

Example 4: Stress-Extension Analysis for Polymer Blends (isotactic polypropylene/SPC-1)

The stress-extension behavior of a blend of isotactic polypropylene (first polymer component) and sample SPC-1 as described in Tables 1-3 (second polymer component) was measured. The procedure used was as follows. A total of 72 g of a mixture of the first polymer component and the second polymer component were mixed in a Brabender intensive mixture for 3 minutes at a temperature controlled to be within the range of 185°C to 220°C. High shear roller blades were used for the mixing, and approximately 0.4 g of Irganox®1076, an antioxidant available from Ciba-Geigy, was added to the blend. At the end of the mixing, the mixture was removed and pressed out into a 6" x 6" (15 cm x 15 cm) mold into a pad 0.25" (6 mm) thick at 215°C for 3 to 5 minutes. At the end of this period, the pad was cooled and removed and allowed to anneal for 3 to 5 days. Test specimens of the required dumbbell geometry were removed from this pad and evaluated on an Instron® tester to produce the data shown in Table 4.

The first polymer component was Escorene® 4292, a commercially available homoisotactic polypropylene from ExxonMobil Chemical Company, Houston, Texas; it is denoted "isotactic polypropylene" or "iPP" in the following

data tables. The second polymer component was SPC-1 as characterized in Tables 1, 2 and 3. Shaded areas represent broken samples. Clear areas represent lack of data due to extension beyond machine limits.

Table 4

| | Composition (g) | | | | |
|---------|-----------------|------|------|------|------|
| iPP | 64 | 56 | 48 | 40 | 32 |
| SPC-1 | 8 | 16 | 24 | 32 | 40 |
| | Stress (psi) | | | | |
| E =10% | 4836 | 4243 | 3839 | 3274 | 2878 |
| E = 25% | 2782 | 3526 | 3460 | 3238 | 2863 |
| E = 50% | 2566 | 2539 | 2472 | 2589 | 2218 |
| E =100% | | 2509 | 2434 | 2231 | 2169 |
| E =150% | | | | 2239 | 2130 |
| E =200% | | | | 2247 | 2105 |
| E =250% | | | | 2245 | 2093 |
| E =300% | | | | 2253 | 2066 |
| E =350% | | | | 2251 | 2073 |
| E =400% | | | | 2251 | 2137 |
| E =450% | | | | 2247 | 2158 |
| E =500% | | | | 2246 | 2177 |
| E =550% | | | | | 2229 |

5

Example 5: Stress-Extension Analysis for Polymer Blends (isotactic polypropylene/SPC-2)

The procedure of Example 4 was repeated, using SPC-2 (as described in Tables 1-3) as the second polymer component. The results are shown in Table 5. Shaded areas represent broken samples. Clear areas represent lack of data due to extension beyond machine limits.

10

Table 5

| | Composition (g) | | | | |
|---------|-----------------|------|------|------|------|
| iPP | 64 | 56 | 48 | 40 | 32 |
| SPC-2 | 8 | 16 | 24 | 32 | 40 |
| | Stress (psi) | | | | |
| E =10% | 4616 | 3477 | 2777 | 2221 | 1405 |
| E = 25% | 2754 | 2863 | 2319 | 2178 | 1518 |
| E = 50% | | 2459 | 2221 | 1911 | 1517 |
| E =100% | | | 2243 | 1872 | 1522 |
| E =150% | | | 2261 | 1910 | 1546 |
| E =200% | | | 2271 | 1947 | 1581 |
| E =250% | | | 2317 | 2037 | 1696 |
| E =300% | | | 2341 | 2061 | 1788 |
| E =350% | | | | 2078 | 1919 |
| E =400% | | | | 2167 | 2096 |
| E =450% | | | | 2221 | 2319 |
| E =500% | | | | 2397 | 2597 |
| E =550% | | | | | 2976 |
| E =600% | | | | | 3611 |

Example 6: Stress-Extension Analysis for Polymer Blends (isotactic polypropylene/SPC-3)

5

The procedure of Example 4 was repeated, using SPC-3 (as described in Tables 1-3) as the second polymer component. The results are shown in Table 6. Shaded areas represent broken samples. Clear areas represent lack of data due to extension beyond machine limits.

Table 6

| | Composition (g) | | | | |
|---------|-----------------|------|------|------|------|
| iPP | 64 | 56 | 48 | 40 | 32 |
| SPC-3 | 8 | 16 | 24 | 32 | 40 |
| | Stress (psi) | | | | |
| E =10% | 3700 | 3333 | 2427 | 1574 | 770 |
| E = 25% | 2614 | 2989 | 2229 | 1607 | 840 |
| E = 50% | | 2428 | 1944 | 1632 | 895 |
| E =100% | | 2399 | 1999 | 1644 | 945 |
| E =150% | | 2405 | 2043 | 1648 | 989 |
| E =200% | | | 1995 | 1653 | 1069 |
| E =250% | | | | | 1140 |
| E =300% | | | | | 1195 |
| E =350% | | | | | 1244 |
| E =400% | | | | | 1273 |

Example 7: Stress-Extension Analysis for Polymer Blends (isotactic polypropylene/SPC-4)

5

The procedure of Example 4 was repeated, using SPC-4 (as described in Tables 1-3) as the second polymer component. The results are shown in Table 7. Shaded areas represent broken samples. Clear areas represent lack of data due to extension beyond machine limits.

Table 7

| | Composition (g) | | | | |
|---------|-----------------|------|------|------|------|
| iPP | 64 | 56 | 48 | 40 | 32 |
| SPC-4 | 8 | 16 | 24 | 32 | 40 |
| | Stress (psi) | | | | |
| E =10% | 4485 | 3719 | 3091 | 2387 | 1879 |
| E = 25% | 3983 | 3467 | 2944 | 2413 | 1960 |
| E = 50% | 2691 | 2473 | 2264 | 2186 | 1818 |
| E =100% | | 2448 | 2390 | 1951 | 1713 |
| E =150% | | 2496 | 2436 | 1965 | 1742 |
| E =200% | | 2523 | 2449 | 2001 | 1775 |
| E =250% | | 2532 | 2456 | 2038 | 1790 |
| E =300% | | 2537 | 2445 | 2070 | 1781 |
| E =350% | | 2548 | 2434 | 2087 | 1765 |
| E =400% | | 2564 | 2467 | 2152 | 1878 |
| E =450% | | 2552 | | 2242 | 1998 |
| E =500% | | | | 2407 | 2255 |
| E =550% | | | | 2642 | 2603 |
| E =600% | | | | 3048 | 3020 |
| E =650% | | | | | 3410 |

Example 8: (Comparative) Stress-Extension Analysis for Polymer Blends
(isotactic polypropylene/EPR)

The procedure of Example 4 was repeated using isotactic polypropylene and conventional EPR as a comparative example. The results are shown in Table 8. Shaded areas represent broken samples.

Table 8

| | Composition (g) | | | | |
|---------|-----------------|------|------|------|------|
| iPP | 64 | 56 | 48 | 40 | 32 |
| EPR | 8 | 16 | 24 | 32 | 40 |
| | Stress (psi) | | | | |
| E =10% | 3456 | 3125 | 2589 | 960 | 789 |
| E = 25% | | 3358 | 3596 | 1122 | 999 |
| E = 50% | | | | 1536 | 1356 |
| E =100% | | | | 2125 | 1681 |
| E =150% | | | | | 1674 |
| E =200% | | | | | 1744 |

Example 9: (Comparative) Stress-Extension Analysis for Polymer Blends
(isotactic polypropylene/aePP)

5 The procedure of Example 4 was repeated using isotactic polypropylene and conventional aePP as a comparative example. The results are shown in Table 9. Shaded areas represent broken samples.

Table 9

| | Composition (g) | | | | |
|---------|-----------------|------|------|------|------|
| iPP | 64 | 56 | 48 | 40 | 32 |
| aePP | 8 | 16 | 24 | 32 | 40 |
| | Stress (psi) | | | | |
| E =10% | 3258 | 3012 | 2258 | 872 | 635 |
| E = 25% | | | 2785 | 1687 | 975 |
| E = 50% | | | | | 1012 |
| E =100% | | | | | 974 |

10 Figure 1 shows the stress-elongation behavior of several samples of polymer blends of an FPC and an SPC. In Figure 1, the curves are labeled with the percent by weight of the SPC in the blend.

For comparison, Figure 2 shows the stress-elongation behavior of a blend of an FPC and EPR; i.e., instead of an SPC according to the present invention, the blend includes conventional EPR (Vistalon® 457, an EP rubber sold by the ExxonMobil Chemical Company, Houston TX). The polymer blends of the present invention show greater elongation than conventional blends.

Figure 3 shows the flexural modulus of blends of the present invention, as a function of the weight percent of the SPC in the FPC/SPC blend. The curves are labeled with the percent by weight of ethylene in the SPC.

For comparison, Figure 4 shows the higher flexural modulus of a blend of an FPC and EPR.

Example 10: Foamed Polymer Blends

Foams were formed from several polymer blends, including blends of the present invention and conventional blends for comparison. All of the foaming tests were conducted at the Polymer Processing Institute (PPI) of the New Jersey Institute of Technology (NJIT), Newark, NJ, under identical operating conditions. A Killion single screw extruder of 1.25" (32 mm) diameter with 40 L/D equipped with a high-pressure gas injection port located at 19 L/D length and with a 1/8" (3.2 mm) rod die was used. Carbon dioxide gas from a compressed cylinder was used at various pressures ranging from 300 to 900 psi. In all the cases, the materials were plasticated within the first 19 diameter. Beyond the gaseous injection port, the remaining extruder length was used for gas mixing, compression and cooling of the gas-laden polymer. At the die exit, the temperature was 160°C, which was kept constant during all the experiments when the foams were made under various CO₂ pressures. This die temperature value was based on various experimental trials on the materials of the following illustrative examples.

The samples tested were as follows:

Sample 10A (comparative):

A commercial isotactic polypropylene product, PP 6523 from Montell, was used. Some of its main characteristics as determined in our labs at the

Baytown Polymer Center are as follows:

MFR ~4;

conventional GPC molecular weights, $M_w \sim 360k$, $M_z \sim 1050k$ and MWD
(M_w/M_n) 15.4;

5 Young's modulus 53,000 psi, yield stress 4200 psi, and elongation at break
940%, all at room temperature at 2'/min by Instron test (equivalent
ASTM method-D524);

DSC melting temperature T_m (2nd heat at the 10°C/min heat rate)
~162.3°C;

10 $T_c \sim 114.2^\circ\text{C}$;

$\Delta H \sim 111.2 \text{ J/g}$.

Sample 10B (comparative):

15 A sample of iPP prepared during the running of the ExxonMobil in-house
pilot plant facility was used as a control sample. The catalyst used was PX-525B-
16 (Q metallocene). The sample characteristics were as follows:

MFR ~2;

$M_w \sim 392k$; $M_z \sim 870k$ and MWD ~8.3;

20 Young's modulus 66,000, yield stress 4900 psi and elongation at break
760%;

$T_m \sim 153^\circ\text{C}$;

$T_c \sim 120.0^\circ\text{C}$; $\Delta H \sim 108.0 \text{ J/g}$.

Sample 10C:

25 The material used for this example was a melt mixed blend of iPP of
Sample 10B and a metallocene-derived EP copolymer ("M-EP") made in our
Vistalon® pilot plant. Typically, these M-EP polymers may have PP content in
the range of 65 to 95 percent by weight. The specific M-EP sample used had PP
content of 88 wt. % and a Mooney viscosity of 16.5. The blend composition was
30 95 wt. % iPP (of Sample 10B) to 5 wt % M-EP. The blend was prepared prior to
foaming. Any conventional plastic-mixing unit such as Braebender or
plasticating/mixing extrusion device can be used to make such blends following

normal PP processing conditions. In this case, a lab size twin mixing screw extruder used for making various blends of desired compositions was used to make the blend. No difficulty in mixing the two components and pelletizing the extrudates was encountered. Blend compositions in the pellet form were used in the Killion extruder using the procedure described above.

Sample 10D:

This sample was the same as Sample 10C, except that the isotactic polypropylene/M-EP ratio was 90/10 by weight.

Figures 5 and 6 show electron micrographs of several foamed samples. Figures 5A and 6A show the prior art foamed sample 10A, with a density of 0.7 g/cm³, Figure 5B shows foamed sample 10C (0.5 g/cm³), and Figure 6B shows sample 10D (0.6 g/cm³). Based on various attempts to make foams of samples 10A-D, clear and distinct advantages in samples 10C and 10D were observed. The advantages were not only in the foam production process, but also in the final foam quality.

At any given (foaming) gas pressure, the formation of foam was easier for sample 10C than for sample 10A. In the case of prior art sample 10A, an excessive amount of bubble collapse was noted in the gas pressure range of 250 to 750 psi. In addition, the foamed product of this material had excessively large bubbles and the texture was poor (see Figures 5A and 6A). The foamed extrudates spiraled excessively and coiled at the die exit due to bubble bursting. Also, it was discovered that the density of the foam material of sample 10A was relatively high, 0.7 g/cm³ in the best of circumstances.

In contrast, the materials of samples 10C and 10D could be foamed with ease. The foam product quality was significantly better in these two samples as well. The foamed extrudates had fine textures, and their bubbles (foam cells) were more uniform and showed practically no bursting upon die exit, as is clearly seen in comparing Figures 5A and 5B, and Figures 6A and 6B. In addition, foams of a broad density range (~0.1 to 0.7 g/cm³) could be successfully made from these materials. It is believed that the advantageous properties observed for samples 10C and 10D will be obtained even in cases where other gases, such as

heptane, nitrogen, helium, butane, etc., are used as blowing agents. Some of these gases are currently used by the polymer processing industry in foaming high polymers such as polystyrene and PET. The solubility and diffusion of gas from the polymer play a role in determining foam properties, and it is believed that these factors will be favorable for the blend compositions of the present invention.

Example 11: Rheological Properties of Polymer Blends

Select rheological properties of several polymer blends were measured. In the following blends, the first polymer component (FPC) is an M-isotactic polypropylene copolymer having an ethylene content of 0.9 wt %, an MFR of 1.0, $M_w \sim 3.6 \times 10^5$, $M_w/M_n \sim 3.2$, $M_z/M_w \sim 2.3$, $M_{z+1}/M_w \sim 3.6$, $J_r \sim 4.5 \times 10^4 \text{ Pa}^{-1}$, and $T_m \sim 149^\circ\text{C}$. The second polymer component (SPC) is an M-EP having 12.4 wt % ethylene and Mooney viscosity ~ 16.5 . The SPC also contained Irganox® and Irgafox (Ciba-Geigy). Table 10 shows the composition of each sample.

Table 10

| Composition | Sample No. | | | | | |
|-------------|------------|-----|-----|-----|-----|------|
| | 11A | 11B | 11C | 11D | 11E | 11F* |
| FPC | 99 | 98 | 95 | 90 | 80 | 100 |
| SPC | 1 | 2 | 5 | 10 | 20 | 0 |

* control

In addition to samples 11A-11F, two commercially available polymer compositions were tested. Sample 11G was Mobil HMS LDPE LFA033B, and sample 11H was Dow Styron® 685D. The values of the measured parameters are shown in Table 11, as a function of shear rate. Viscosities were measured at 200°C . Die swell was measured as described above.

Table 11

| | Shear Rate (s ⁻¹) | | | | | | | |
|-------------------|-------------------------------|---------|---------|---------|---------|---------|---------|---------|
| | 20 | 50 | 80 | 100 | 200 | 500 | 800 | 1000 |
| Sample 11A | | | | | | | | |
| Shear Stress (Pa) | 65950.2 | 92818.8 | 112360 | 122130 | 155105 | 216170 | 255252 | 273571 |
| Die Swell | 1.56 | 1.74 | 1.79 | 1.75 | | | | |
| Viscosity (Pa•s) | 3290.14 | 1856.49 | 1405.39 | 1221.38 | 775.575 | 432.368 | 319.085 | 273.557 |
| Sample 11B | | | | | | | | |
| Shear Stress (Pa) | 56179.8 | 86712.3 | 103810 | 117245 | 151441 | 208842 | 244260 | 262579 |
| Die Swell | 1.57 | 1.71 | 1.75 | 1.79 | | | | |
| Viscosity (Pa•s) | 2802.71 | 1734.36 | 1298.46 | 1172.52 | 757.254 | 417.711 | 305.344 | 262.566 |
| Sample 11C | | | | | | | | |
| Shear Stress (Pa) | 51294.6 | 76941.9 | 98925.3 | 107474 | 145335 | 207621 | 243039 | 257694 |
| Die Swell | 1.53 | 1.68 | 1.73 | 1.74 | | | | |
| Viscosity (Pa•s) | | | | | | | | |
| Sample 11D | | | | | | | | |
| Shear Stress (Pa) | 58622.4 | 87933.6 | 107474 | 117245 | 150220 | 212506 | 243039 | 265022 |
| Die Swell | 1.54 | 1.66 | 1.78 | 1.82 | | | | |
| Viscosity (Pa•s) | | | | | | | | |
| Sample 11E | | | | | | | | |
| Shear Stress (Pa) | 20762.1 | 50073.3 | 72056.7 | 76941.9 | 118466 | 159990 | | |
| Die Swell | 1.67 | 1.73 | 1.82 | 1.82 | 1.98 | | | |
| Viscosity (Pa•s) | 1035.78 | 1001.53 | 901.285 | 769.468 | 592.368 | 320.001 | | |
| Sample 11F | | | | | | | | |
| Shear Stress (Pa) | 68392.8 | 96482.7 | 117245 | 127245 | 155105 | 217391 | 254030 | 274792 |
| Die Swell | 1.63 | 1.77 | 1.83 | 1.82 | | | | |
| Viscosity (Pa•s) | 3412 | 1929.78 | 1466.5 | 1270.23 | 775.575 | 434.81 | 317.558 | 274.778 |
| Sample 11G | | | | | | | | |
| Shear Stress (Pa) | 29311.2 | 58622.4 | 76941.9 | 90376.2 | 138007 | 218613 | 260137 | 273571 |
| Die Swell | 1.55 | 1.85 | 1.91 | 2.1 | 2.17 | | | |
| Viscosity (Pa•s) | 1462.28 | 1172.52 | 962.389 | 903.819 | 690.078 | 437.253 | 325.192 | 273.557 |
| Sample 11H | | | | | | | | |
| Shear Stress (Pa) | 76941.9 | 118466 | 146556 | 155105 | 201514 | 268686 | 311431 | 311431 |
| Die Swell | 1.47 | 1.79 | 1.86 | 1.92 | | | | |
| Viscosity (Pa•s) | 3838.5 | 2369.47 | 1833.12 | 1551.15 | 1007.64 | 537.406 | 389.314 | 311.415 |

The data are shown graphically in Figures 7A and 7B. Figure 7A shows die swell versus shear stress for samples 11A-F, and Figure 7B shows similar data for the conventional samples 11G and 11H for comparison.

While the illustrative embodiments of the invention have been described with particularity, it will be understood that various other modifications will be apparent to and can be readily made by those skilled in the art without departing from the spirit and scope of the invention. Accordingly, it is not intended that the scope of the claims appended hereto be limited to the examples and descriptions set forth herein but rather that the claims be construed as encompassing all the features of patentable novelty which reside in the present invention, including all features which would be treated as equivalents thereof by those skilled in the art to which the invention pertains.

All documents cited herein are fully incorporated by reference for all jurisdictions in which such incorporation is permitted and to the extent they are not inconsistent with this specification. All documents to which priority is claimed are fully incorporated by reference for all jurisdictions in which such incorporation is permitted. Although dependent claims have single dependencies in accordance with U.S. practice, each of the features in any of the dependent claims can be combined with each of the features of one or more of the other dependent claims dependent upon the same independent claim or claims.

CLAIMS

1. A foamed polymer composition comprising a foamed blend of:
 - (a) a first polymer component having a melting point greater than 110°C and comprising:
 - (i) polypropylene, or
 - (ii) a copolymer of propylene and at least one comonomer selected from the group consisting of ethylene and C₄–C₁₂ α-olefins, the copolymer having a comonomer content of less than 10% by weight; and
 - (b) a second polymer component comprising at least one random copolymer of propylene and at least one comonomer selected from the group consisting of ethylene and C₄–C₈ α-olefins, the second polymer component having crystallinity of from 2% to 65% derived from isotactic polypropylene sequences, a propylene content of from 65% to 95% by weight, and a melting point of from 25°C to 105°C,the first polymer component being present in an amount by weight of from 50% to 99.9%, based on the total weight of the first and second polymer components.
2. The composition of claim 1, wherein the first polymer component has a melting point greater than 115°C.
3. The composition of claim 1, wherein the first polymer component has a melting point greater than 130°C.
4. The composition of claim 1, wherein the comonomer of the first polymer component is ethylene.
5. The composition of claim 1, wherein the comonomer content of the first polymer component is less than 8% by weight.

6. The composition of claim 1, wherein the comonomer content of the first polymer component is less than 5% by weight.
- 5 7. The composition of claim 1, wherein the comonomer of the second polymer component is ethylene.
8. The composition of claim 1, wherein the propylene content of the second polymer component is from 70% to 92.5% by weight.
- 10 9. The composition of claim 1, wherein the propylene content of the second polymer component is from 75% to 90% by weight.
- 15 10. The composition of claim 1, wherein the propylene content of the second polymer component is from 80% to 89% by weight.
11. The composition of claim 1, wherein the second polymer component has a crystallinity of from 2% to 50%.
- 20 12. The composition of claim 1, wherein the second polymer component has a crystallinity of from 5% to 45%.
13. The composition of claim 1, wherein the second polymer component has a crystallinity of from 10% to 40%.
- 25 14. The composition of claim 1, wherein the first polymer component is present in an amount by weight of from 70% to 99.5%, based on the total weight of the first and second polymer components.
- 30 15. The composition of claim 1, wherein the first polymer component is present in an amount by weight of from 80% to 99%, based on the total weight of the first and second polymer components.

16. The composition of claim 1, wherein the second polymer component comprises a mixture of a first copolymer of propylene and ethylene or a C₄–C₈ α-olefin, and a second copolymer of propylene and ethylene or a C₄–C₈ α-olefin.
17. The composition of claim 1, wherein the second polymer component comprises a mixture of a first copolymer of propylene and ethylene and a second copolymer of propylene and ethylene.
18. The composition of claim 1, wherein the second polymer component comprises a mixture of a first copolymer of propylene and ethylene having an ethylene content of 5% to 9% by weight and a second copolymer of propylene and ethylene having an ethylene content of 10% to 22% by weight.
19. The composition of claim 1, wherein the composition has a density of 0.3 g/cm³ to 0.7 g/cm³.
20. The composition of claim 1, wherein the composition has a density of 0.4 g/cm³ to 0.6 g/cm³.
21. An article comprising the foamed polymeric composition of claim 1.
22. A foamed polymer composition consisting essentially of a foamed blend of:
- (a) a first polymer component having a melting point greater than 110°C and comprising:
 - (i) polypropylene, or
 - (ii) a copolymer of propylene and ethylene, the copolymer having an ethylene content of less than 10% by weight; and

(b) a second polymer component comprising at least one random copolymer of propylene and ethylene, the second polymer component having crystallinity of from 2% to 65% derived from isotactic polypropylene sequences, a propylene content of from 65% to 95% by weight, and a melting point of from 25°C to 105°C, the first polymer component being present in an amount by weight of from 50% to 99.9%, based on the total weight of the first and second polymer components.

23. The composition of claim 22, wherein the first polymer component has a melting point greater than 115°C.

24. The composition of claim 22, wherein the first polymer component has a melting point greater than 130°C.

25. The composition of claim 22, wherein the comonomer content of the first polymer component is less than 8% by weight.

26. The composition of claim 22, wherein the comonomer content of the first polymer component is less than 5% by weight.

27. The composition of claim 22, wherein the propylene content of the second polymer component is from 70% to 92.5% by weight.

28. The composition of claim 22, wherein the propylene content of the second polymer component is from 75% to 90% by weight.

29. The composition of claim 22, wherein the propylene content of the second polymer component is from 80% to 89% by weight.

30. The composition of claim 22, wherein the second polymer component has a crystallinity of from 2% to 50%.

31. The composition of claim 22, wherein the second polymer component has a crystallinity of from 5% to 45%.

5 32. The composition of claim 22, wherein the second polymer component has a crystallinity of from 10% to 40%.

33. The composition of claim 22, wherein the first polymer component is present in an amount by weight of from 70% to 99.5%, based on the total
10 weight of the first and second polymer components.

34. The composition of claim 22, wherein the first polymer component is present in an amount by weight of from 80% to 99%, based on the total weight of the first and second polymer components.

15 35. The composition of claim 22, wherein the second polymer component comprises a mixture of a first copolymer of propylene and ethylene and a second copolymer of propylene and ethylene.

20 36. The composition of claim 22, wherein the second polymer component comprises a mixture of a first copolymer of propylene and ethylene having an ethylene content of 5% to 9% by weight and a second copolymer of propylene and ethylene having an ethylene content of 10% to 22% by weight.

25 37. The composition of claim 22, wherein the composition has a density of 0.3 g/cm³ to 0.7 g/cm³.

30 38. The composition of claim 22, wherein the composition has a density of 0.4 g/cm³ to 0.6 g/cm³.

39. An article comprising the foamed polymeric composition of claim 22.

40. A process for preparing a foamed polymer composition, comprising:
- (a) providing a first polymer component having a melting point greater than 110°C and comprising:
 - (i) polypropylene, or
 - (ii) a copolymer of propylene and at least one comonomer selected from the group consisting of ethylene and C₄–C₁₂ α-olefins, the copolymer having a comonomer content of less than 10% by weight;
 - (b) providing a second polymer component comprising at least one random copolymer of propylene and at least one comonomer selected from the group consisting of ethylene and C₄–C₈ α-olefins, the second polymer component having crystallinity of from 2% to 65% derived from isotactic polypropylene sequences, a propylene content of from 65% to 95% by weight, and a melting point of from 25°C to 105°C;
 - (c) blending the first and second polymer components in amounts such that the first polymer component is present in an amount by weight of from 50% to 99.9%, based on the total weight of the first and second polymer components; and
 - (d) contacting the blended polymer components with a foaming agent sufficient to produce a foamed polymer composition.
41. The process of claim 40, wherein the foaming agent comprises carbon dioxide, nitrogen, a noble gas, a hydrocarbon gas, a carbon dioxide gas releasing agent or a nitrogen gas releasing agent.
42. The process of claim 40, wherein the step of contacting the blended polymer components with a foaming agent is carried out so that the foamed polymer composition has a density of from 0.3 g/cm³ to 0.7 g/cm³.
43. A foamed polymer composition produced by the method of claim 40.

44. A process for preparing a foamed polymer composition, comprising:
- (a) providing a first polymer component having a melting point greater than 110°C and comprising:
 - (i) polypropylene, or
 - (ii) a copolymer of propylene and ethylene, the copolymer having a comonomer content of less than 10% by weight;
 - (b) providing a second polymer component comprising at least one random copolymer of propylene and ethylene, the second polymer component having crystallinity of from 2% to 65% derived from isotactic polypropylene sequences, a propylene content of from 65% to 95% by weight, and a melting point of from 25°C to 105°C;
 - (c) blending the first and second polymer components in amounts such that the first polymer component is present in an amount by weight of from 50% to 99.9%, based on the total weight of the first and second polymer components; and
 - (d) contacting the blended polymer components with a foaming agent sufficient to produce a foamed polymer composition.
45. The process of claim 44, wherein the foaming agent comprises carbon dioxide, nitrogen, a noble gas, a hydrocarbon gas, a carbon dioxide gas releasing agent or a nitrogen gas releasing agent.
46. The process of claim 44, wherein the step of contacting the blended polymer components with a foaming agent is carried out so that the foamed polymer composition has a density of from 0.3 g/cm³ to 0.7 g/cm³.
47. A foamed polymer composition produced by the method of claim 44.

EP CLAIMS

48. Foamed polymer composition comprising a foamed blend of:
- (a) a first polymer component having a melting point greater than 110°C and comprising:

- (i) polypropylene, or
- (ii) a copolymer of propylene and at least one comonomer selected from the group consisting of ethylene and C₄-C₁₂ α-olefins, the copolymer having a comonomer content of less than 10% by weight;

- (b) a second polymer component comprising at least one random copolymer of propylene and at least one comonomer selected from the group consisting of ethylene and C₄-C₈ α-olefins, the second polymer component having crystallinity of from 2% to 65% derived from isotactic polypropylene sequences, a propylene content of from 65% to 95% by weight, and a melting point of from 25°C to 105°C;

the first polymer component being present in an amount by weight of from 50% to 99.9%, based on the total weight of the first and second polymer components.

49. Composition of claim 48, wherein the first polymer component has a melting point greater than 115°C and preferably greater than 130°C.

50. The composition of claim 48 or claim 49, wherein the comonomer content of the first polymer component is less than 8% by weight, and preferably less than 5% by weight.

51. Composition according to any of claims 48-50, wherein the propylene content of the second polymer component is from 70% to 92.5% by weight, preferably from 75% to 90% by weight, and most preferably from 80 to 89% by weight.

52. Composition according to any of claims 48-51, wherein the second polymer component has a crystallinity of from 2% to 50%, preferably from 5% to 45% and most preferably from 10% to 40%.

53. Composition according to any of claims 48-52, wherein the first polymer component is present in an amount by weight of from 70% to 99.5%, based on the total weight of the first and second polymer components, preferably in an amount by weight of from 80% to 99%, based on the total weight of the first and second polymer components.
54. Composition according to any of claims 48-53, wherein both polymer components are copolymers.
55. Composition according to any of claims 48-54, wherein both components are copolymers of propylene and ethylene.
56. Composition according to any of claims 48-55, wherein the second polymer component comprises a mixture of a first copolymer of propylene and ethylene having an ethylene content of 5% to 9% by weight and a second copolymer of propylene and ethylene having an ethylene content of 10% to 22% by weight.
57. Composition according to any of claims 48-56, wherein the composition has a density of 0.3 g/cm^3 to 0.7 g/cm^3 , preferably of 0.4 g/cm^3 to 0.6 g/cm^3 .
58. An article comprising a foamed composition according to any of the claims 48-57.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US01/32298

A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) : C08J 9/00; C08L 23/00, 23/04

US CL : 521/59, 134, 525/240

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 521/59, 134; 525/240

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No |
|-----------|---|----------------------|
| X | US 6,040,348 A (DELAITE et al) 21 March 2000, col. 2, line 6-51, col. 4 line 48-50, col. 5 line 59-67, examples 1, 2, claims 1-14 | 1-58 |
| A | US 4,870,114 A (HASHIMOTO et al) 26 September 1989, entire document | 1-58 |

☐ Further documents are listed in the continuation of Box C.
 ☐ See patent family annex.

| | |
|---|--|
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07 FEBRUARY 2002

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